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Many-Body Effects in Semiconductor Quantum Dots

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1 Introduction

In recent years, semiconductor quantum dots (QDs) have been studied extensively due to possible applications in optoelectronic devices like LEDs, lasers, or amplifiers[1,2]. In the rapidly emerging field of quantum information technology, QDs have been successfully used to demonstrate the generation of single photons or correlated photon pairs[3–5]. Furthermore, the strong coupling regime for QD emitters in optical microcavities has been demonstrated[6,7]. A common aspect in fundamental studies and practical applications of QDs is the critical role of correlation and scattering processes of carriers which are studied within this project.

Optical studies of QDs have been recently focused on self-assembled systems which are typically grown in the Stranski-Krastanoff mode. The resulting QDs are randomly distributed on a two-dimensional wetting layer (WL). The energy spectrum of this system consists of discrete states which correspond to a three-dimensional carrier localization in the QDs and a quasi-continuum of states at higher energies in connection with the two-dimensional motion of carriers within the WL. Scattering processes of carriers between the localized QD states as well as between localized and delocalized states are possible due to the Coulomb interaction in addition to carrier-phonon interaction.

2 Quantum Kinetics of Carrier-Phonon Interaction

At low carrier densities and elevated temperatures the interaction of carriers with LO-phonons provides the dominant contribution to scattering channels for redistributing carriers in QDs as well as to the dephasing of optical excitations. Due to the discrete nature of the QD energy spectra, a phonon bottleneck has been predicted by energy conservation arguments based on Fermi's golden rule: if the energy spacing between QD states does not exactly match the LO-phonon energy $\hbar\omega_{\mathrm{LO}}$, carrier scattering would not be possible in this picture. The phonon-bottleneck problem is still a debated topic because there is experimental evidence for[8,9] as well as against it[10,11]. Recently it has been discussed, that a perturbational treatment of the carrier-phonon interaction based on Fermi's golden rule is not applicable to discrete QD systems.

Because of the strong interaction between carriers and phonons in QDs, a theoretical description has to be based on polarons[12,13]. The physical picture of a polaron as a carrier in a crystal interacting with a surrounding cloud of lattice distortions is shown in Fig. 1a. The energy spectrum of polarons differs significantly from that of free carriers as shown in

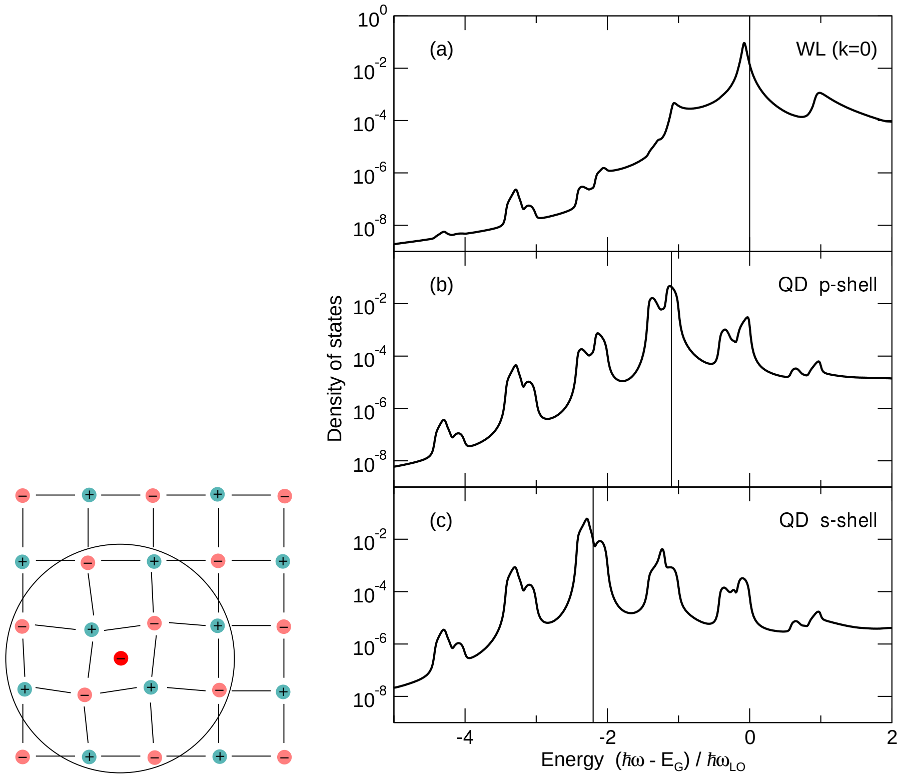

Figure 1. Electron interacting with surrounding cloud of lattice distortions (left) and density of states (DOS) for a polaron at the bandedge a), in the p-shell b) and in the s-shell c) (right). Vertical lines indicate the δ-like DOS of free electrons.

Fig. 1. There and in the following examples we consider QDs with two confined shells for electrons and holes. For the assumed cylindrical symmetry they are called s- and p-shell where the latter is two-fold degenerate in addition to the spin degeneracy[14]. In the polaron picture, energy renormalizations that incorporate shifts and broadening of the electronic states as well as phonon satellites and hybridization effects immediately invalidate the simple arguments for the phonon bottleneck.

To study the experimentally observed fast carrier relaxation in QDs on the level of a microscopic theory, we solve numerically the coupled quantum-kinetic equations for the carrier and polarization dynamics. These equations incorporate high-dimensional scattering integrals for the carrier-phonon interaction as well as memory integrals over the history of the system[14, 15].

The scattering rate between polaron states is proportional to the overlap of the density-of-states (DOS) of the involved states, shifted by one phonon energy against each other. From Fig. 1 one immediately finds that in the free particle picture scattering between QD states only is possible if the level-spacing exactly matches the LO-phonon energy due to the δ-like DOS. This corresponds to the prediction of the phonon bottleneck. In the polaron picture even for large detunings between the level spacing and the LO-phonon energy, sufficient overlap is present which gives rise to fast carrier scattering.

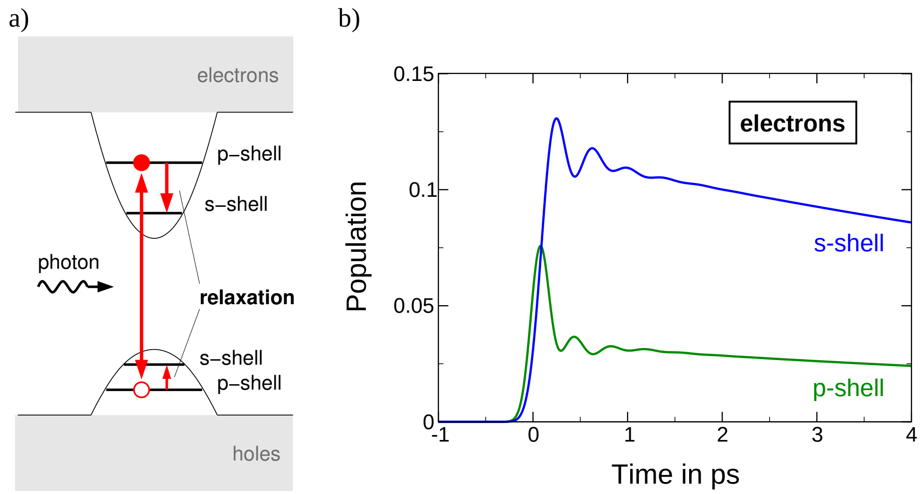

Figure 2. Resonant optical excitation condition for the semiconductor QD system a) and corresponding temporal evolution of the population probability for the QD states. Calculations are done for InGaAs/GaAs QDs at 300K and a detuning of 10% between level spacing and phonon energy.

In the following example we consider the situation where a resonantly tuned laser pulse excites carriers only in the p-shell and the subsequent relaxation of carriers into the s-shell is studied, see Fig. 2a. The corresponding evolution of the carrier population is shown in Fig. 2b. First the p-shell is populated due to pulse excitation which is followed by a fast relaxation into the s-shell. Rabi-oscillations due to memory effects indicate the strong coupling regime for the carrier-phonon interaction. At later times a decrease of the QD population is observed due to escape of carriers into the WL, which is also included in the calculation.

Our results show that even for materials with weak polar coupling, like InGaAs, QD polarons lead to fast carrier scattering on a sub-picosecond timescale which is influenced only weakly by the level spacing[14].

3 Quantum Kinetics of Carrier-Carrier Interaction

For optoelectronic applications of semiconductor nano-structures the optical gain spectra of the active material are of central importance. Under the assumption that the carrier system is in a thermodynamic quasi-equilibrium, we evaluate the temperature and carrier-density dependence of the optical gain spectra theoretically. To achieve this, the knowledge of the many-body effects is of central importance, because the line-broadening as well as the lineshape of the optical spectra is governed by dephasing and correlation processes[16]. At room temperatures and elevated carrier densities the important dephasing mechanisms are the carrier-carrier Coulomb interaction as well as the interaction of carriers with LO phonons.

For the case of a high excitation density, the screening of the Coulomb interaction in the coupled QD-WL system justifies a treatment of the carrier-carrier scattering in the second-order Born approximation while the carrier-phonon interaction is included in the random phase approximation (RPA). It turns out that in the coupled QD-WL system the Markov

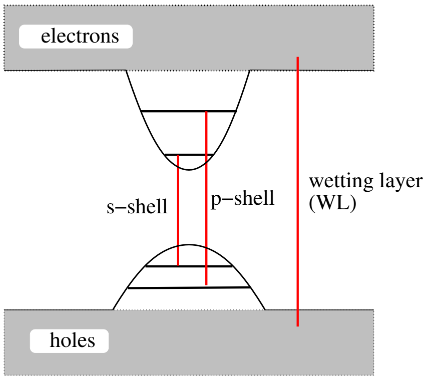

Figure 3. Shell structure of the combined QD-WL system showing localized QD states for electrons (holes) which are energetically below a quasi-continuum of delocalized WL states corresponding to the free motion in the WL. The red lines show the allowed optical transitions.

approximation cannot be applied and renormalized quasi-particle properties (which in the case of low carrier densities are the polarons) need to be included for a proper description of the optical spectra[17]. In Fig. 3 the electronic structure of the discussed system is schematically shown. We consider two confined shells for electrons as well as for holes so that from the allowed transitions (red lines in Fig. 3) we expect three resonances in the optical spectra, namely the ground state resonance, the excited state resonance and the excitonic resonance of the WL.

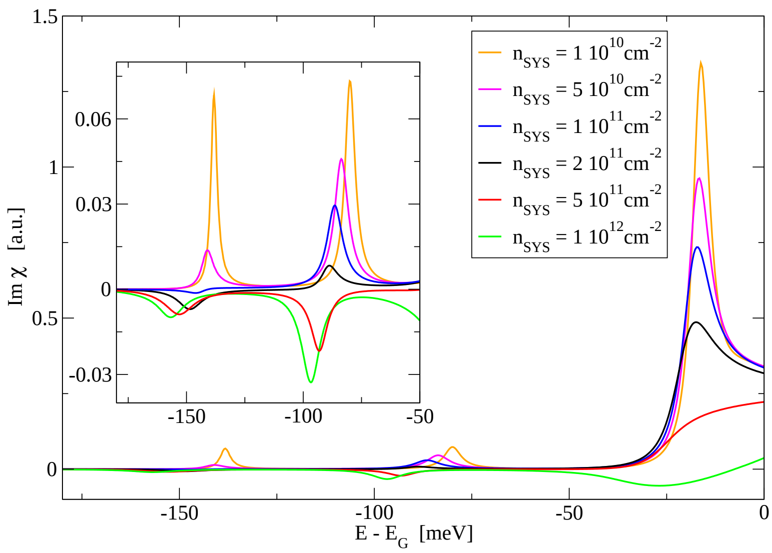

Figure 4. Optical absorption spectra for the combined QD-WL system including interaction-induced dephasing and line shifts due to Coulomb interaction and carrier-phonon interaction for various total carrier densities. The inset shows a scale up of the QD resonances.
Calculations are done for InGaAs/GaAs QDs at 300K.

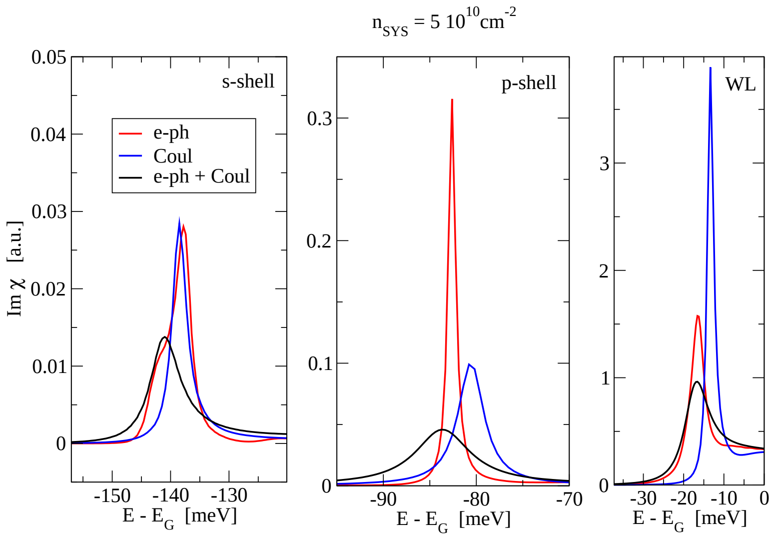

Figure 5. Influence of carrier-carrier and carrier-phonon interaction on the optical absorption spectra. Given are the energetic region of the ground state resonance, excited state resonance, and excitonic resonance of the WL. The red (blue) line is a calculation with carrier-phonon (carrier-carrier) interaction only, while the black line shows the full calculation.
Calculations are done for InGaAs/GaAs QDs at 300K.

In Fig. 4 the results of the calculations are presented. Given is the absorption vs. energy (relative to the band-gap energy E_G). We obtain the mentioned three resonances at around -130meV,-85meV, and -15meV, respectively. With increasing carrier density the resonances are bleached out, reach transparency, and switch to negative absorption (optical gain). Additionally observed is a distinct, correlation-induced red-shift of the QD resonances with increasing carrier-density, which has also been found in recent photoluminescence experiments[18].

In Fig. 5 we compare the role of carrier-carrier and carrier-phonon interaction for a fixed carrier density. We find that both mechanisms are equally important to properly describe the broadening of all three resonances even at intermediate to high excitation conditions.

4 Summary

A quantum-kinetic treatment for carrier-phonon and carrier-carrier interaction in semiconductor quantum dots has been used to analyze the efficiency of scattering processes and their influence on the carrier-density dependent optical spectra. The absence of a phonon bottleneck is explained in terms of polaron renormalizations. The combined influence of the scattering processes is responsible for the observed dephasing and line-shift effects in the optical gain spectra.

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References

1. Y. Masumoto and T. Takagahara, eds., *Semiconductor Quantum Dots* (Springer-Verlag, Berlin, 2002), 1st ed.
2. P. Michler, ed., *Single Quantum Dots* (Springer-Verlag, Berlin, 2003), 1st ed.
3. P. Michler, A. Imamoglu, M. D. Mason, P. J. Carson, G. F. Strouse, and S. K. Buratto, Nature **406**, 968 (2000).
4. E. Moreau, I. Robert, L. Manin, V. Thierry-Mieg, J. M. Gerad, and I. Abram, Phys. Rev. Lett. **87**, 183601 (2001).
5. M. Pelton, C. Santori, J. Vuckovic, B. Zhang, G. S. Solomon, J. Plant, and Y. Yamamoto, Phys. Rev. Lett. **89**, 233602 (2002).
6. J. P. Reithmaier, G. Sek, A. Löffler, C. Hofmann, S. Kuhn, S. Reitzenstein, L. V. Keldysh, V. D. Kulakovskii, T. L. Reinecke, and A. Forchel, Nature **432**, 197 (2004).
7. T. Yoshie, A. Scherer, J. Hendrickson, G. Khitrova, H. M. Gibbs, G. Rupper, C. Ell, O. B. Shchekin, and D. G. Deppe, Nature **432**, 200 (2004).
8. J. Urayama, T.B. Norris, J. Singh, and P. Bhattacharya. Observation of phonon bottleneck in quantum dot electronic relaxation. *Phys. Rev. Lett.*, **86**:4930, 2001.
9. W.W.E. Minnaert, A.Yu. Silov, W. van der Vleuten, J.E.M. Haverkort, and J.H. Wolter. Fröhlich interaction in InAs/GaAs self-assembled quantum dots. *Phys. Rev. B*, **63**:75303, 2001.
10. E. Tsitsishvili, R.v. Baltz, and H. Kalt. Temperature dependence of polarization relaxation in semiconductor quantum dots. *Phys. Rev. B*, **66**:R161405, 2002.
11. E. Péronne, F. Fossard, F.H. Julien, J. Brault, M. Gendry, B. Salem, G. Bremond, and A. Alexandrou. Dynamic saturation of an intersublevel transition in self-organized InAs/$In_xAl_{1-x}As$ quantum dots. *Phys. Rev. B*, **67**:205329, 2003.
12. T. Inoshita and H. Sakaki. Density of states and phonon-induced relaxation of electrons in semiconductor quantum dots. *Phys. Rev. B*, **56**:R4355, 1997.
13. O. Verzelen, R. Ferreira, G. Bastard, T. Inoshita, and H. Sakaki. Polaron effects in quantum dots. *phys. stat. sol. (a)*, **190**:213, 2002.
14. J. Seebeck, T.R. Nielsen, P. Gartner, and F. Jahnke. Polarons in semiconductor quantum dots and their role in the quantum kinetics of carrier relaxation. *Phys. Rev. B*, **71**:125327, 2005.
15. J. Seebeck, T.R. Nielsen, P. Gartner, and F. Jahnke. Quantum kinetic theory of phonon-assisted carrier transitions in nitride-based quantum-dot systems. *arXiv:cond-mat/cond-mat/0509692*, 2005.
16. H. Haug and S.W. Koch. *Quantum Theory of the Optical and Electronic Properties of Semiconductors*, World Scientifiv Publ., Singapore, 4. edition, 2004.
17. M. Lorke, T. R. Nielsen, J. Seebeck, P. Gartner, and F. Jahnke , Influence of carrier-carrier and electron-phonon correlations on optical absorption and gain in quantum-dot systems, *arXiv.org:cond-mat/0509543*, 2005.
18. K. Matsuda, K. Ikeda, T. Saiki, H. Saito, K. Nishi Carrier-carrier interaction in single $In_{0.5}Ga_{0.5}As$ quantum dots at room temperature investigated by near-field scanning optical microscope, Appl. Phys. Lett., **83**, 2250 (2003).